

### Eechnical Mote

Mo. 322

# SURFACE TENSIONS OF NORMAL AND PARA HYDROGEN

R. J. CORRUCCINI



U. S. DEPARTMENT OF COMMERCE NATIONAL BUREAU OF STANDARDS

#### THE NATIONAL BUREAU OF STANDARDS

The National Bureau of Standards is a principal focal point in the Federal Government for assuring maximum application of the physical and engineering sciences to the advancement of technology in industry and commerce. Its responsibilities include development and maintenance of the national standards of measurement, and the provisions of means for making measurements consistent with those standards; determination of physical constants and properties of materials; development of methods for testing materials, mechanisms, and structures, and making such tests as may be necessary, particularly for government agencies; cooperation in the establishment of standard practices for incorporation in codes and specifications; advisory service to government agencies on scientific and technical problems; invention and development of devices to serve special needs of the Government; assistance to industry, business, and consumers in the development and acceptance of commercial standards and simplified trade practice recommendations; administration of programs in cooperation with United States business groups and standards organizations for the development of international standards of practice; and maintenance of a clearinghouse for the collection and dissemination of scientific, technical, and engineering information. The scope of the Bureau's activities is suggested in the following listing of its four Institutes and their organizational units.

Institute for Basic Standards. Applied Mathematics. Electricity. Metrology. Mechanics. Heat. Atomic Physics. Physical Chemistry. Laboratory Astrophysics.\* Radiation Physics. Radio Standards Laboratory: Radio Standards Physics; Radio Standards Engineering. Office of Standard Reference Data.

Institute for Materials Research. Analytical Chemistry. Polymers. Metallurgy. Inorganic Materials. Reactor Radiations. Cryogenics.\* Materials Evaluation Laboratory. Office of Standard Reference Materials.

Institute for Applied Technology. Building Research. Information Technology. Performance Test Development. Electronic Instrumentation. Textile and Apparel Technology Center. Technical Analysis. Office of Weights and Measures. Office of Engineering Standards. Office of Invention and Innovation. Office of Technical Resources. Clearinghouse for Federal Scientific and Technical Information.\*\*

Central Radio Propagation Laboratory.\* Ionospheric Telecommunications. Tropospheric Telecommunications. Space Environment Forecasting. Aeronomy.

<sup>\*</sup> Located at Boulder, Colorado 80301.

<sup>\*\*</sup> Located at 5285 Port Royal Road, Springfield, Virginia 22171.

## NATIONAL BUREAU OF STANDARDS Technical Mote 322

ISSUED August 11, 1965

#### SURFACE TENSIONS OF NORMAL AND PARA HYDROGEN

R. J. Corruccini Institute for Materials Research National Bureau of Standards Boulder, Colorado

NBS Technical Notes are designed to supplement the Bureau's regular publications program. They provide a means for making available scientific data that are of transient or limited interest. Technical Notes may be listed or referred to in the open literature.

#### CONTENTS

		F	Page
ΑE	STRAC	CT	1
1.	INTR	ODUCTION	1
2.	DISCU	USSION OF THE EXPERIMENTAL RESEARCHES	
	AND	THE CORRECTIONS OF THE DATA	2
	2.1.	Kamerlingh Onnes and Kuypers	2
	2.2.	Van Itterbeek	4
		Table 1 - Original and Corrected Data from	
		Two Older Sources	5
	2.3.	Grigor'ev	6
		Table 2 - Data by Grigor'ev [1964] Calculated	
		from Eqs. (5) and (6)	7
3.	COMF	PARISON OF THE CORRECTED DATA	8
4.	A FUI	NCTION TO FIT THE DATA	8
	Table	e 3 - Smoothed Values of Surface Tension as	
	Giver	n by Eqs. (8) and (9) 1	0
5.	APPE	NDIX I. Relation to Other Correlations 1	1
6.	APPE	NDIX II. Contact Angle 1	2
7.	ACKN	OWLEDGMENT 1	3
8.	REFE	RENCES 1	4
Fig	gure 1.	Schematic drawing of capillary rise method.  h' is the capillary rise (calculated) of the annular meniscus in the reservoir above a hypothetical infinite reservoir	ó
Fig	gure 2.	Corrected experimental surface tension points of Kamerlingh Onnes and Kuypers and of Van Itterbeek and straight lines representing the experimental points of Grigor'ev 17	

#### Surface Tensions of Normal and Para Hydrogen

#### R. J. Corruccini

The published data on the surface tensions of normal hydrogen from three investigations are apparently in disagreement by about 10%. In this note it is shown that these data are actually in agreement within about 1% if properly computed. The resulting concordant data are represented by functions and by a table suitable for linear interpolation.

Key Words: Hydrogen, Liquid, Surface tension

#### 1. Introduction

Until recently, data on the surface tension of hydrogen were available from only two experimental researches, namely, those of Kamerlingh Onnes and Kuypers [1914] and Van Itterbeek [1940]. These appeared to be in good agreement. However, in a paper on helium, Van Urk, Keesom, and Kamerlingh Onnes [1925] stated in a footnote that a particular meniscus correction had not been properly applied by Kamerlingh Onnes and Kuypers. They gave recalculated values for hydrogen which were about 10% lower and which thus disagree by about this much with Van Itterbeek's later values. Some compilers [e.g., Friedman 1954] have been unaware of the recalculation by Van Urk et al., and others have chosen to ignore it [e.g., Stewart, Germann, and McCarty, 1962]. Some have applied corrections like those of Van Urk et al. to the data of Kamerlingh Onnes and Kuypers [e.g., Landolt-Bornstein Tabellen and the International Critical Tables], but the large differences between the results so obtained and Van Itterbeek's data were not accounted for. Thus there has been no clear agreement in the literature on a set of "best" values for this property.

Recently new measurements have appeared. Grigor'ev and Rudenko [1964] have determined normal hydrogen, and Grigor'ev [1964] determined parahydrogen. (It must be assumed that the measurements by Kamerlingh Onnes and Kuypers and Van Itterbeek were made on normal hydrogen.) This recent work agrees well with the corrected data of

Van Urk et al. but not with Van Itterbeek's data.

In this note the application of the meniscus corrections in the older investigations is examined, and it is shown that Van Itterbeek's data, if properly corrected, in fact agree well with both Grigor'ev's and those of Kamerlingh Onnes and Kuypers as corrected by Van Urk et al. All of the available experimental data are thus shown to be in close accord. The resulting corrected data are represented by functions and by a table suitable for linear interpolation.

#### Discussion of the Experimental Researches and the Corrections of the Data

#### 2.1. Kamerlingh Onnes and Kuypers

These authors measured the rise, h, in a capillary situated on the axis of a tubular reservoir. The reservoir tube had an inside diameter of only about 1 cm, and hence the liquid in this reservoir itself had an appreciable capillary rise, h', which had to be computed and added to the observed rise in the capillary tube. Figure 1 shows the situation and the notation used.

In an earlier work with similar apparatus but other fluids, Verschaffelt [1895] had measured the height, d, of the annular meniscus and had found it to be less than if the cross section of this meniscus in an axial plane were semicircular. Hence he derived a formula for h' on the assumption that the cross section was a semi-ellipse of semi-minor axis, d, and major axis, R-r<sub>1</sub>:

$$h' = \left(h + \frac{r}{3}\right) \left(\frac{2d/(R-r_1)^2}{1/r - 2d/(R-r_1)^2}\right). \tag{1}$$

This formula was used by Kamerlingh Onnes and Kuypers, but since they were unable to measure d, they made the simplest available, but probably incorrect, assumption, namely, that the cross section was semicircular, so that  $2d = R-r_1$ . (There is a misprint in their statement of this equation, as well as in their value of r, which should have read 0.03316 cm rather than 0.3316 cm; thus compare r/3 as given in their Table II. Their calculations were apparently not affected by these

misprints.)

There is a further small and well-established correction for the meniscus in the capillary tube. This, to a good approximation, has the value r/3 [Adam 1941]. Hence the corrected capillary rise is

$$H = h + h' + r/3$$
. (2)

This is the hypothetical rise above a reference surface of zero curvature; i.e., above an infinite reservoir. It is related to the surface tension,  $\gamma$ , by the well established formula (see any text on surface chemistry)

$$\gamma = grH \left(\rho_{i} - \rho_{v}\right) / 2 \tag{3}$$

in which g is the acceleration of gravity,  $\rho_{\ell}$  is the density of the liquid, and  $\rho_{v}$  is the density of the vapor above the meniscus.

Later, Verschaffelt [1921] re-examined the question of the shape of the annular meniscus by a graphical construction and published tables from which d and h' could be found, given the values of h and the three radii, r, r<sub>1</sub>, and R. This procedure was an improvement, since it avoided arbitrary assumption of too simple a meniscus shape.

The recalculation by Van Urk et al. used Verschaffelt's 1921 tables, and their language gives the impression that this was the sole change. However, inspection shows that this produced only a 3% reduction in H. Most of the roughly 10% reduction in Y resulted from correction of a different error which was not specifically mentioned. Thus, in place of eq. (3), Kamerlingh Onnes and Kuypers had used the following formula:

$$\gamma = gH(\rho_{\ell} - \rho_{v})/2\left(\frac{1}{r} - \frac{1}{R-r_{1}}\right). \tag{4}$$

The origin of this formula was not indicated. However, one can see that it differs from eq. (3) by the term  $1/(R-r_1)$ , which is evidently a correction for the curvature of the annular meniscus. But such a correction is

already contained in h'. Hence eq. (4) applies it a second time. The meaning of the various corrections is best seen by referring back to Verschaffelt [1895] who was the only one to give an explanatory discussion. However, eq. (4) is not to be found in his paper.

In table 1 are listed the data of Kamerlingh Onnes and Kuypers and the corrections by Van Urk et al. In table 1 two further adjustments are shown which are small: (1) The temperature scale has been adjusted from the n-H<sub>2</sub> vapor-pressure scale of Kamerlingh Onnes and Keesom [1913] to that of Woolley, Scott, and Brickwedde [1948], and thence to the NBS 1955 scale, and (2) more modern density data recommended by Stewart and Roder [1964] have been used. The temperature of the lowest point given by Van Urk et al. was variously reported as 14.78° (Kamerlingh Onnes and Kuypers, table I), 14.66° (Kamerlingh Onnes and Kuypers, table II), and 14.68° (Van Urk et al.). Because of this ambiguity, this point has been omitted from consideration.

#### 2.2. Van Itterbeek

The experimental method was the same as that of Kamerlingh Onnes and Kuypers, and the surface tension was calculated using the same eqs. (1), (2), and (4). The only difference was that Van Itterbeek measured the meniscus height, d. Kamerlingh Onnes and Kuypers had been unable to do this. Van Itterbeek was aware of the use of Verschaffelt's tables of 1921 by Van Urk et al. He used these tables to obtain a value of d but found it to be about twice as large as his experimental value (and, incidentally, about the same as if the meniscus section were semicircular). One must infer that he thought that this discrepancy cast doubt on Verschaffelt's 1921 tables since he proceeded to calculate h' from Verschaffelt's ellipse formula, eq. (1), inserting his measured value of d.

However, analysis shows that it makes little difference which method is used to obtain h' in this case, as the resulting H values differ by only about 2%. It is not easy to make a choice. The correction given by eq. (1) has the advantage of using the experimental and thus presumably true value of d, but has the disadvantage that the assumed elliptical meniscus shape may be incorrect. In contrast, Verschaffelt's table undoubtedly gives a more correct representation of the curvature of the meniscus at its bottom, upon which h' depends directly. The unfortunate fact that it yields too large a value of d could result from non-zero contact angle, inasmuch as the derived d (but not h') would be evidently very sensitive to contact angle at angles near zero. (However, see Appendix II.) I have chosen to obtain h' from Verschaffelt's table.

Table 1

Original and Corrected Data from Two Older Sources

	<b>.</b>	dynes/cm		2.634	2,317	2. 196 <sub>5</sub>	1.910		2.393	1.956		
nis Note	۵	g/cm <sup>3</sup>			0,00035	. 00067	. 00085	.00136		.00061	.00132	
As corrected in this Note	P	g/cm <sup>3</sup>		0.07524	.07345	.07271	.07084		.07375	96010.		
As cor	H	cm		Values	of VanUrk	et al. adopted			1,697	1.457		
	, H ,	(NBS 1955)	14]	16. 198	18,050	18.764	20.453		17.747	20.354		
cted by x et al.	>	dynes/cm	Kamerlingh Onnes and Kuypers [1914]	2, 631	2.319 <sub>5</sub>	2. 194 <sub>5</sub>	1.910	[1940]				
As corrected by Van Urk et al.	н	cm	nnes and ]	2, 169	1.962	1,883	1.694	Van Itterbeek [1940]				
	¥	dynes/cm	nerlingh C	2.919	2.575	2.438	2. 12 <sub>6</sub>	Van	2.66	2. 19		
	م <sup>۵</sup>	g/cm <sup>3</sup> c	Kar	0.0003	. 0005	. 0008	.0013		a.0061	. 0013		
As originally reported	p	g/cm <sup>3</sup>		0.0751	.07335	. 0726	. 0708		. 0740	. 0708		
s original	Н	cm		2.231	2,021	1.941	1.749		1.658	1.447		
A	त	cm		2.064	1.869	1.794	1.616	ř	1.540 <sub>5</sub>	1.323 1.447		
	[ <del>-</del>	»,		16.16	17.99	18.70	20.40		17.72	20.32	,	

an error. Should be 0.00061

A much larger correction of about 12% results from using eq. (3) rather than eq. (4). This is the principal adjustment made here to Van Itterbeek's results. It brings his data into good agreement with the corrected Leiden values. The data are listed in table 1. Since Van Itterbeek listed the vapor pressures at his experimental points, it was possible to adjust his temperatures and densities to the same basis to which I have adjusted the Leiden data. These adjustments are small, as can be seen in table 1.\*

#### 2.3. Grigor'ev

The difference in rise in two adjacent capillaries of different diameters was measured. This technique eliminates the correction h'. As in most Russian papers the details are glossed over and the data points appear only in a small graph. Nevertheless, the conventional technique inspires confidence. The author's estimate of accuracy was  $\sim 0.5\%$ . For the first time both normal and parahydrogen were determined.

The data were determined from about 17° to 20.4°K and were represented by the following linear equations:

$$n - H_2$$
:  $Y = 5.25 - 0.162 T$  (5)

$$p - H_2$$
:  $Y = 5.27 - 0.165 T$ . (6)

Several errors have been noted in Van Itterbeek's table I: (1) A misprint of  $\rho_{\rm V}$  at 17.72°K which should read 0.00061. (2) The two values of h'. The second value is evidently a misprint. If rearranged from 0.0146 to 0.1046, which is essentially the result obtained by repeating his calculation, then it is possible to confirm his value of H, otherwise not. (3) The value of H at 20.32° cannot be accounted for by using either his erroneous value of h' or the value 0.0939 cm obtained by following his method of calculation.

However, these extrapolate to zero at a temperature roughly one degree below critical, whereas physical reality requires the surface tension to vanish at the critical point. The temperature scale on which the results are given is not defined; nor are the density data explicitly given, but the usage in an earlier paper [Grigor'ev 1963] suggests that the data of Woolley, Scott, and Brickwedde were used for the liquid densities and If this were the case, adjustthe ideal gas law for the vapor densities. ments might be made to real gas densities and to more modern density data for liquid parahydrogen [Stewart and Roder], but the net change would not exceed 0.1%. If Grigor'ev's temperatures were based on the hydrogen vapor pressure data of Woolley, Scott, and Brickwedde, no adjustment larger than 0.005 degree would be called for. If, on the other hand, he were on the Russian P.R.M.I. scale, recent comparisons have shown this to be as much as 0.03 degree higher than the NBS 1955 scale in the range under consideration. This would correspond to a difference in surface tension of 0.005 dyne/cm or about 0.2%. Inasmuch as there is no clear basis for making any particular corrections and the probable ones discussed above are smaller than Grigor'ev's claimed accuracy of  $\sim 0.5\%$ , no adjustments were made in this note. Also, rather than attempt to extract the data points from the graph in Grigor'ev's paper, I have used values calculated from his equations at the integral temperatures, 17, 18, 19, and 20°K, as if they were original data points. These are given in Table 2.

Table 2

Data by Grigor'ev [1964] Calculated from Eqs. (5) and (6)

Т	<b>Y</b>			
	n-H <sub>2</sub>	p-H <sub>2</sub>		
۰K	dynes/cm	dynes/cm		
17	2.496	2.465		
18	2.334	2.300		
19	2.172	2.135		
20	2.010	1.970		

#### 3. Comparison of the Corrected Data

The corrected data from table 1 and the two linear equations of Grigor'ev are plotted in figure 2. The spread in the data for  $n-H_2$  at any given temperature is not more than 1.3%. This is regarded as good agreement for data taken over a span of fifty years. The  $n-H_2$  minus  $p-H_2$  difference as given by Grigor'ev ranges from 1 to 2%.

The four Leiden points are accurately linear. However, both this set and the data of Van Itterbeek extrapolate to zero at appreciably below the critical temperature, thus demonstrating again the inadequacy of the linear formula over a wide range.

#### 4. A Function to Fit the Data

The simplest representation is a linear function of temperature, as was used by Grigor'ev. If fitted to the data, it represents them within the experimental precision, but is not reliable for extrapolation. A straight line passed through the experimental points and constrained to zero at the critical temperature provides a better wide-range representation, but deviates systematically from the line fitted to only the experimental points by up to 1%. To put it another way, the slopes would differ by at least 5%. Thus, use of a more elaborate function is justified.

We will adopt the empirical equation of Van der Waals [Güggenheim 1950]:

$$\gamma = \gamma_0 (1 - T_r)^n , \qquad (7)$$

in which T<sub>r</sub> is the reduced temperature and n is a constant, which for many classical substances has been found to have the value 11/9. On fitting this separately to the four sets of data in tables 1 and 2, the following parameters were found:

Investigation		n —	Υ <u>ο</u>
Kamerlingh Onnes : and Kuypers	n - H <sub>2</sub>	1.116	5.565
Van Itterbeek :	n - H <sub>2</sub>	1.08	5.479
Grigor'ev :	n - H <sub>2</sub>	1.05	5.329
Grigor'ev :	p - H <sub>2</sub>	1.07	5.372

In view of the near constancy of n for other substances, it is reasonable to adopt a fixed value for both forms of hydrogen and let only  $\gamma_{\text{O}}$  be a fitted parameter. Evidently n should be roughly 1.1 rather than 11/9. The decreased value of n is a quantum effect [Hirschfelder et al. 1954. A further decision must be made: namely, how to weight the three investigations of n - H2. However this is to be done, it seems desirable to preserve the normal vs para differences found by Grigor'ev, inasmuch as these differences may well be more accurate than either set of data alone due to partial cancellation of the systematic errors that we may suppose to have been present. These differences could be preserved in various ways, e.g., by averaging the three sets of data on n - H2 and subtracting from this the normal minus para differences by Grigor'ev. It would also be reasonable to adopt only Grigor'ev's data for both forms, inasmuch as his experimental method is better established, and his curve for n - H2 is seen to closely approximate the older data as corrected here. The latter course has been adopted. We assume an average n of 1.065 which leads to new values of  $\gamma_o$  of 5.369 (n-H<sub>2</sub>) and 5.328 (p-H<sub>2</sub>). Thus the equations have the following equivalent forms:

$$n - H_2 \left\{ \begin{array}{c} \gamma = 5.369 \left(1 - T_r\right)^{1.065} \\ \log \gamma = 0.729865 + 1.065 \log \left(1 - 0.0301386T\right) \end{array} \right\}$$
 (8)

$$p - H_2 \left\{ \begin{array}{c} \gamma = 5.328 \left(1 - T_r\right)^{1.065} \\ \log \gamma = 0.72653 + 1.065 \log \left(1 - 0.0303251T\right) \end{array} \right\}. \quad (9)$$

The temperature reducing parameters used were as follows [Stewart and Roder]:

$$n - H_2$$
:  $T_c = 33.18$ ° K  
 $p - H_2$ :  $T_c = 32.976$ ° K.

These formulas are within  $\pm 0.2\%$  of the equations of Grigor'ev in the range of his experimental data. They deviate from the four points of Kamerlingh Onnes and Kuypers by +0.6% (avg.) and +1.3% (max.) and deviate from the two points of Van Itterbeek by -0.3% and -0.7%. In table 3 are presented values at integral temperatures calculated from

Table 3
Smoothed Values of Surface Tension as Given by Eqs. (8) and (9)

T	<b>Y</b>		T	<b>Y</b>	
	n-H <sub>2</sub>	p-H <sub>2</sub>		n-H <sub>2</sub>	p-H <sub>2</sub>
°K	dynes/cm	dynes/cm	°K	dynes/cm	dynes/cm
13.803 (t.p.)	-	2.990	23	1.525	1.491
13.947 (t.p.)	3.004	-	24	1.366	1.333
14	2.995	2.958	25	1.208	1. 175
15	2.829	2.792	26	1.052	1.019
16	2.663	2.627	27	.896	.864
17	2.498	2.462	28	.743	.711
18	2.334	2.298	29	. 591	. 560
19	2.171	2. 135	30	.442	.411
20	2.008	1.973	31	. 296	. 266
20.268 (n.b.p.	.) -	1.930	32	. 154	. 125
			en jeden de service de Service de service de		
20.38 (n.b.p.	.) 1. 946	-	32.976 (c.p.)	-	0
21	1.847	1.812	33.18 (c.p.)	0	
22	1.686	1.651			

eqs. (8) and (9). Up to 32°K linear interpolation in this table will introduce no errors larger than one in the last place.

#### 5. Appendix I. Relation to Other Correlations

<u>Friedman</u> [1954] adopted the original data of Kamerlingh Onnes and Kuypers and Van Itterbeek. He was apparently unaware of the corrections by Van Urk et al. He represented the combined data by a linear equation not constrained at the critical point.

Chelton and Mann [1956] referenced Friedman and repeated his equation. The original sources were not referenced.

The Arthur D. Little Hydrogen Handbook [Anon., 1960] referenced Chelton and Mann and repeated the above equation. The earlier sources in this sequence were not referenced. Thus, starting from this Handbook, it is necessary to go through the two previous compilations in order to ascertain the original sources of the data.

The CEL-WADD Compendium [Johnson, 1960] states that "the data of Kamerlingh Onnes and Kuypers appears to be the only available to date." It presents a table purporting to be from that source. However, the data and reference were actually taken from the obsolete International Critical Tables (1928). The ICT compiler was the same Verschaffelt already mentioned. Evidently he corrected the data of Kamerlingh Onnes and Kuypers for presentation in the ICT, though without specifically saying that he had done so. The ICT data are close to the corrected values by Van Urk et al., though not identical with them.

Landolt-Bornstein Tabellen. The 6th edition has sections on surface tension and contact angle in Vol. II, Part 3 (1956). These contain no data on hydrogen. This is strange because the 5th edition [1931] gives smoothed values based on Kamerlingh Onnes and Kuypers but corrected by Verschaffelt and de Block. These are very close to the corrected values by Van Urk et al.

Stewart, Germann, and McCarty [1962]. This work employed a quantum-mechanical corresponding states correlation of the available data on the isotopic species of hydrogen and helium. The input data on hydrogen were the original data of Kamerlingh Onnes and Kuypers, the corrected data by Van Urk et al., and the data of Van Itterbeek. The data by Kamerlingh Onnes and Kuypers and by Van Urk et al. were treated as independent investigations. The authors adopted the uncorrected data of Kamerlingh Onnes and Kuypers and of Van Itterbeek in preference to the corrected data by Van Urk et al. because the former were concordant.

In summary, the various experimental data, compilations, and correlations may be seen to fall into two sets, the second set being of the order of 10% higher than the first set from  $14^\circ$  to  $20^\circ$ K, but the agreement within either set being of the order of  $\pm 1\%$ .

#### Set 1.

- a) Data of Kamerlingh Onnes and Kuypers as corrected by Van Urk et al. or by Verschaffelt (ICT, CEL-WADD Compendium, and Landolt-Bornstein) or by this note.
- b) Data of Van Itterbeek as corrected by this note.
- c) Data of Grigor'ev.
- d) This note.

#### Set 2.

- a) Uncorrected data of Kamerlingh Onnes and Kuypers.
- b) Uncorrected data of Van Itterbeek.
- c) Correlation by Friedman of (a) and (b). Reproduction in Cryogenic Data Book and Arthur D. Little Hydrogen Handbook.
- d) Correlation by Stewart, Germann, and McCarty.

#### 6. Appendix II. Contact Angle

The meniscus correction methods applied to all of the experimental data that we have cited rely on the assumption that the contact angle between the liquid hydrogen and the wall is zero. None of the experimenters on surface tension observed the contact angle.

Recently workers at Arthur D. Little, Inc. [1961] reported measurements of contact angles between liquid hydrogen and various solids, including several metals and Teflon. These measurements were made by observing a cross section of the meniscus on a round rod sample with a protractor telemicroscope. The results varied from 0° to 90° and were not consistent with each other or with reasonable expectations based on the surface energies of the substances involved.

Later Good and Ferry [1963] experimented with the same substances. They observed the angle of normal reflections from the meniscus at the contact zone and found zero contact angle in all cases. They pointed out that the method used at ADL readily gives apparent non-zero angles if the telemicroscope is not focused exactly in the plane containing the rod axis.

In view of the precision, self-consistency, and reasonableness of the results of Good and Ferry, the application of meniscus corrections based on the assumption of zero contact angle was evidently proper.

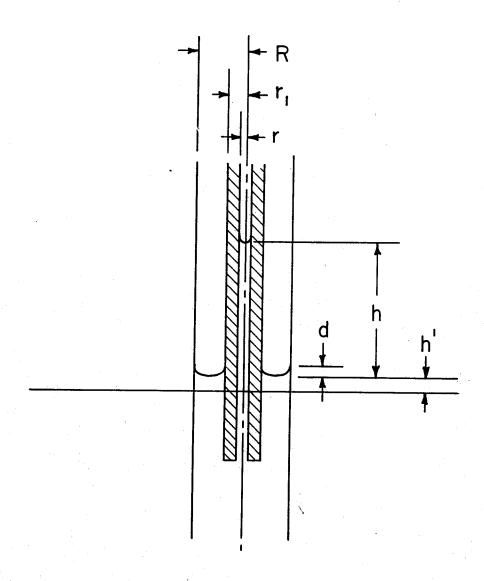
#### 7. Acknowledgment

I am most appreciative of a painstaking review of this paper by Dr. J. M. H. Levelt Sengers.

#### 8. References

- Adam, N. K. (1941), The Physics and Chemistry of Surfaces, 3rd ed., pp. 363-371. (Oxford University Press, Oxford).
- Anon. (1960), Hydrogen Handbook, Arthur D. Little, Inc., AFFTC TR-60-19.
- Arthur D. Little, Inc. (Nov. 1961), Report No. 63270-00-03, pp. 93-96, NASA Contract No. NAS5-664.
- Chelton, D. B., and D. B. Mann (1956), Cryogenic Data Book, UCRL-3421.
- Friedman, A. S. (1954), The Thermodynamic and Transport Properties of Liquid Hydrogen and Its Isotopes. Part II. Unpublished.
- Good, R. J., and G. V. Ferry (1963), Advances in Cryogenic Engineering 8, 306. (Plenum Press, Inc., New York, N.Y.).
- Grigor'ev, V. N. (1963), Zh. Eksperim. i. Teor. Fiz. 45, 98: translation in Soviet Physics JETP 18, 72 (1964).
- Grigor'ev, V. N. (1964), Zh. Eksperim. i. Teor. Fiz. 47, 484: translation in Soviet Physics JETP 20, 321 (1965).
- Grigor'ev, V. N., and N. S. Rudenko (1964), Zh. Eksperim. i. Teor. Fiz. 47, 92: translation in Soviet Physics JETP 20, 63 (1965).
- Guggenheim, E. A. (1950), Thermodynamics pp. 167-169. (North Holland, Amsterdam/Interscience, New York, N.Y.).
- Hirschfelder, J. O., C. F. Curtiss, and R. B. Bird (1954), Molecular Theory of Gases and Liquids, p. 430, Fig. 6.6-4 (John Wiley and Sons, New York, N.Y./Chapman and Hall, London).
- Johnson, V. J., ed. (1960), A Compendium of the Properties of Materials at Low Temperatures (Phase I). Part I. p. 9.002, WADD Tech. Report 60-56.
- Kamerlingh Onnes, H., and W. H. Keesom (1913), Commun. Kamerlingh Onnes Lab, Univ. Leiden, 137d.

- Kamerlingh Onnes, H., and H. A. Kuypers (1914), Commun. Kamerlingh Onnes Lab, Univ. Leiden, 142d.
- Landolt-Bornstein Phys.-Chem. Tabellen (1931), 5th ed., Suppl. 2, Vol. 1, p. 166.
- Stewart, R. B., F. E. E. Germann, and R. D. McCarty (1962), The Surface Tension of Hydrogen. Unpublished.
- Stewart, R. B., and H. M. Roder (1964), Chapter 11 in Technology and Uses of Liquid Hydrogen. Scott, Denton, and Nicholls, eds. (Pergamon Press, Oxford).
- Van Itterbeek, A. (1940), Physica 7, 325.
- Van Urk, A. Th., W. H. Keesom, and H. Kamerlingh Onnes (1925), Commun. Kamerlingh Onnes Lab, Univ. Leiden, 179a.
- Verschaffelt, J. (1895), Commun. Kamerlingh Onnes Lab, Univ. Leiden, 18.
- Verschaffelt, J. E. (1921), Bull. Classe Sci. Acad. Roy. Belg. 7, 574.
- Woolley, H. W., R. B. Scott, and F. G. Brickwedde (1948), J. Research NBS, 41, 379.



	r	r <sub>i</sub>	R
	(cm)	_ (cm)	(cm)
Onnes - Kuypers	0.03316	0.0801	0.554
<u>Van Itterbeek</u>	.0393	.3264	.6635

Figure 1. Schematic drawing of capillary rise method. h' is the capillary rise (calculated) of the annular meniscus in the reservoir above a hypothetical infinite reservoir.

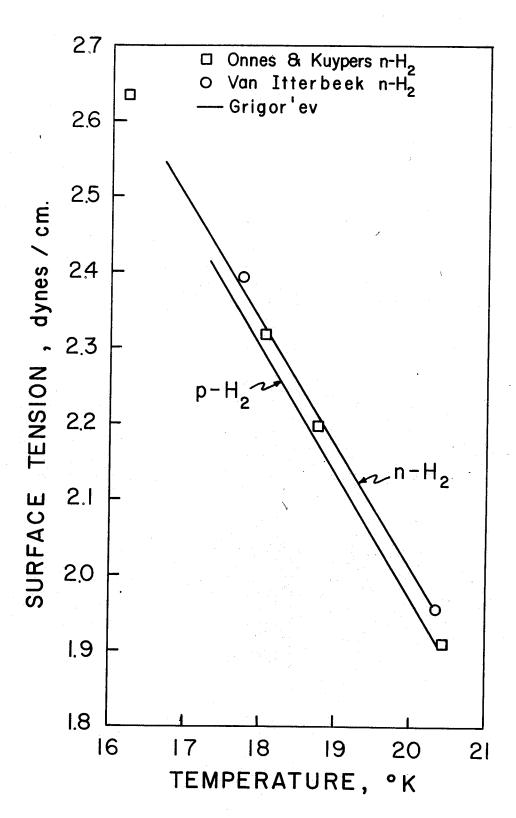


Figure 2.

Corrected experimental surface tension points of Kamerlingh
Onnes and Kuypers and of Van Itterbeek and straight lines representing the experimental points of Grigor'ev.